Supporting Information for

Binary Cu-Co Catalysts Derived from Hydrotalcites with Excellent Activity and Recyclability towards NH₃BH₃ Dehydrogenation

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Figure S1. The experimental setup for hydrogen generation from the NH₃BH₃ dehydrogenation.
**Figure S2.** XRD patterns of the (Cu\textsubscript{x}Co\textsubscript{y})\textsubscript{2}Al-MM0 samples with various ratios of \(x(\text{Cu}) : y(\text{Co})\): a) 1:0, b) 0.87:0.13, c) 0.75:0.25, d) 0.50:0.50, e) 0.25:0.75, f) 0.13:0.87, g) 0:1.

**Figure S3.** SEM images of the MMO samples: A) Cu\textsubscript{2}Al-MM0, B) (Cu\textsubscript{0.50}Co\textsubscript{0.50})\textsubscript{2}Al-MM0, C) Co\textsubscript{2}Al-MM0.
**Figure S4.** Plot of H₂ volume generated from AB hydrolysis vs. time catalyzed by (a) (Cu₀.₅₀Co₀.₅₀)₂Al-300, (b) (Cu₀.₅₀Co₀.₅₀)₂Al-400, (c) (Cu₀.₅₀Co₀.₅₀)₂Al-500, and (d) (Cu₀.₅₀Co₀.₅₀)₂Al-600 (w_{cat} = 20 mg, [AB] = 50 mM, (Cu+Co)/AB=0.09, T = 25±1 °C).

**Figure S5.** XRD patterns of the (Cu₀.₅₀Co₀.₅₀)₂Al-Cat samples obtained by reduction in H₂ at different temperatures: (a) 300 °C, (b) 400 °C, (c) 500 °C, (d) 600 °C.
**Figure S6.** The recyclability for the \( (\text{Cu}_{0.50}\text{Co}_{0.50})_{2}\text{Al-Cat} \) powdered catalyst towards \( \text{NH}_3\text{BH}_3 \) dehydrogenation in three consecutive cycles from curve a to c.

**Figure S7.** SEM-EDS analysis for the CuCoAl-LDHs film precursor on Al substrate prepared by the *in situ* growth method.